

FORM PTO-1390 (Modified) (REV 11-98)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER <b>JMYT-217US</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5) To be assigned <b>09/601694</b>
INTERNATIONAL APPLICATION NO. <b>PCT/GB99/00292</b>	INTERNATIONAL FILING DATE <b>28 January 1999 (28.01.99)</b>	PRIORITY DATE CLAIMED <b>6 February 1998 (06.02.98)</b>	
TITLE OF INVENTION <b>IMPROVEMENTS IN CATALYTIC REDUCTION OF NOx</b>			
APPLICANT(S) FOR DO/EO/US <b>Anders ANDREASSON, Guy Richard CHANDLER, Claus Friedrich GOERSMANN, and James Patrick WARREN</b>			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li>7. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210).</li> <li>8. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>9. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>10. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).</li> <li>11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409).</li> <li>12. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).</li> </ol>			
Items 13 to 20 below concern document(s) or information included:			
<ol style="list-style-type: none"> <li>13. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>15. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li>16. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>17. <input type="checkbox"/> A substitute specification.</li> <li>18. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>19. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail</li> <li>20. <input type="checkbox"/> Other items or information:</li> </ol>			

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5) <div style="font-size: 24pt; font-weight: bold; text-align: center;">09/601694</div>		INTERNATIONAL APPLICATION NO. <div style="font-weight: bold; text-align: center;">PCT/GB99/00292</div>		ATTORNEY'S DOCKET NUMBER <div style="font-weight: bold; text-align: center;">JMYT-217US</div>	
---	--	---	--	--	--

21. The following fees are submitted: <b>BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5) ) :</b> <ul style="list-style-type: none"> <li><input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$970.00</li> <li><input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$840.00</li> <li><input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$690.00</li> <li><input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$670.00</li> <li><input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$96.00</li> </ul> <div style="text-align: center; font-weight: bold; margin-top: 10px;">             ENTER APPROPRIATE BASIC FEE AMOUNT =         </div>				CALCULATIONS    PTO USE ONLY	
				\$840.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <div style="float: right;"> <input type="checkbox"/> 20    <input type="checkbox"/> 30         </div>				\$0.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	12    - 20 =	0	x \$18.00	\$0.00	
Independent claims	3    - 3 =	0	x \$78.00	\$0.00	
Multiple Dependent Claims (check if applicable). <div style="float: right;"><input type="checkbox"/></div>				\$0.00	
TOTAL OF ABOVE CALCULATIONS =				\$840.00	
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <div style="float: right;"><input type="checkbox"/></div>				\$0.00	
SUBTOTAL =				\$840.00	
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <div style="float: right;"> <input type="checkbox"/> 20    <input type="checkbox"/> 30         </div>				\$0.00	
TOTAL NATIONAL FEE =				\$840.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <div style="float: right;"><input type="checkbox"/></div>				\$0.00	
TOTAL FEES ENCLOSED =				\$840.00	
				Amount to be refunded	\$
				charged	\$

☒ A check in the amount of **\$840.00** to cover the above fees is enclosed.

☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.  
A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-0350** A duplicate copy of this sheet is enclosed.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

**SEND ALL CORRESPONDENCE TO:**

**Paul F. Prestia  
Ratner & Prestia  
Suite 301, One Westlakes (Berwyn)  
P.O. Box 980  
Valley Forge, PA 19482**

**Phone: (610) 407-0700**  
**Fax: (610) 407-0701**

**SIGNATURE**

**Christopher R. Lewis**

NAME \_\_\_\_\_

**36,201**

REGISTRATION NUMBER

**August 7, 2000**

DATE \_\_\_\_\_

JMYT-217US

PATENT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Anders Andreasson et al. : Art Unit:  
Application No.: 09/601,694 : Examiner:  
Filed: August 7, 2000 :  
FOR: IMPROVEMENTS IN CATALYTIC :  
REDUCTION OF NO<sub>x</sub> :

SUPPLEMENTAL PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application  
as follows.

IN THE SPECIFICATION:

On page 1, after the title, please insert the following sentence:

--This application is the U.S. national phase application of  
International Application No. PCT/GB99/00292.--

Respectfully submitted,



Christopher R. Lewis, Reg. No. 36,201  
Attorney for Applicants

CRL/lrb

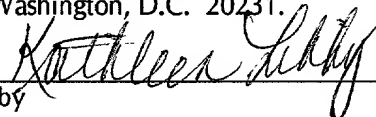
Dated: January 9, 2001

Suite 301  
One Westlakes, Berwyn  
P.O. Box 980  
Valley Forge, PA 19482-0980  
(610) 407-0700

The Assistant Commissioner for Patents is  
hereby authorized to charge payment to  
Deposit Account No. 18-0350 of any fees  
associated with this communication.

EXPRESS MAIL Mailing Label No.: EL736965403US  
Date of Deposit: January 9, 2001

I hereby certify that this paper and fee are being  
deposited, under 37 C.F.R. § 1.10 and with sufficient  
postage, using the "Express Mail Post Office to  
Addressee" service of the United States Postal  
Service on the date indicated above and that the  
deposit is addressed to the Assistant Commissioner  
for Patents, Washington, D.C. 20231.

  
Kathleen Libby

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Anders Andreasson et al. : Art Unit:  
Serial No.: To be Assigned : Examiner:  
Filed: : Herewith :  
FOR: : IMPROVEMENTS IN CATALYTIC :  
REDUCTION OF NO<sub>x</sub> :

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents

Washington, D.C. 20231

Box PCT

S I R :

Prior to examination, please amend the above-identified application  
as follows.

IN THE CLAIMS:

Please amend the following claims:

1 1. (Amended) An improved SCR system for treating combustion  
2 exhaust gas containing [NO] NO<sub>x</sub> and particulates, comprising [in combination and  
3 in or,] an oxidation catalyst effective to convert at least a portion of NO in said  
4 NO<sub>x</sub> to NO<sub>2</sub> thereby enhancing [and enhance] the NO<sub>2</sub> content of the exhaust gas,  
5 a particulate trap, a source of reductant fluid, [injections] injection means for [such]  
6 said reductant fluid located downstream of said particulate trap and an SCR  
7 catalyst.

1 3. (Amended) An SCR system according to claim 1 [or 2], wherein  
2 the oxidation catalyst is a platinum catalyst carried on a through-flow honeycomb  
3 support.

1 4. (Amended) An SCR system according to [claims 1,2 or 3] claim  
2 1, wherein the particulate filter is a wall-flow filter.

1                   5. (Amended) An SCR system according to [any one of the  
2 preceding claims] claim 1, further comprising [also] means to cool gases upstream  
3 of the SCR catalyst.

1                   6. (Amended) An SCR system according to claim 5, further  
2 comprising [also] control means such that said gas cooling means is activated only  
3 when a high SCR catalyst temperature is detected or conditions are determined that  
4 are expected to lead to high catalyst temperatures.

1                   7. (Amended) A diesel engine provided with an SCR system  
2 [according to any one of claims 1 to 5] for treating combustion exhaust gas  
3 containing NO<sub>x</sub> and particulates, said SCR system comprising an oxidation catalyst  
4 effective to convert at least a portion of NO in said NO<sub>x</sub> to NO<sub>2</sub> thereby enhancing  
5 the NO<sub>2</sub> content of the exhaust gas, a particulate trap, a source of reductant fluid,  
6 injection means for said reductant fluid located downstream of said particulate trap  
7 and an SCR catalyst.

1                   8. (Amended) A [light duty] diesel engine according to claim [6] 7,  
2 wherein the volume of the exhaust gas after-treatment system is reduced and the  
3 diesel engine is light duty.

1                   9. (Amended) A method of reducing pollutants, including  
2 particulates and NO<sub>x</sub>, in a gas [streams] stream, comprising passing [such] said gas  
3 stream over an oxidation catalyst under conditions effective to convert at least a  
4 portion of NO in the gas stream to NO<sub>2</sub> [and enhance] thereby enhancing the NO<sub>2</sub>  
5 content of the gas stream, removing at least a portion of said particulates in a  
6 particulate trap, reacting trapped particulate with NO<sub>2</sub>, adding reductant fluid to the  
7 gas stream to form a gas mixture downstream of said trap, and passing the gas  
8 mixture over an SCR catalyst under NO<sub>x</sub> reduction conditions.

1                   11. (Amended) A method according to claims 9 [or 10], wherein  
2 the [gases are] gas stream or gas mixture is cooled[, if necessary,] before reaching  
3 the SCR catalyst.

1                   12. (Amended) A method according to claim 9[, 10 or 11], wherein  
2     the NO to NO<sub>2</sub> ratio of the [gases] gas mixture is adjusted to a level pre-determined  
3     to be optimum for the SCR catalyst, by oxidation of NO over [an] said oxidation  
4     catalyst.

REMARKS

The Assistant Commissioner is hereby authorized to charge payment  
to Deposit Account No. 18-0350 of any fees associated with this communication.

Respectfully submitted,



\_\_\_\_\_  
Paul F. Prestia, Reg. No. 23,031  
Christopher R. Lewis, Reg. No. 36,201  
Attorney for Applicants

/bgd

Dated: August 7, 2000

Suite 301  
1 Westlakes, Berwyn  
P.O. Box 980  
Valley Forge, PA 19482  
(610) 407-0700

**EXPRESS MAIL Mailing Label Number:** EL541609846US  
**Date of Deposit:** August 7, 2000

I hereby certify that this paper and fee are being deposited, under 37 C.F.R. § 1.10 and with sufficient postage, using the "Express Mail Post Office to Addressee" service of the United States Postal Service on the date indicated above and that the deposit is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.



\_\_\_\_\_  
Kathleen Libby

IMPROVEMENTS IN CATALYTIC REDUCTION OF NO<sub>x</sub>

5 The present invention concerns improvements in selective catalytic reduction of NO<sub>x</sub> in waste gas streams such as diesel engine exhausts or other lean exhaust gases such as from gasoline direct injection (GDI).

10 EP 0 758 713 (Toyota) describes a method for purifying the exhaust gas of a diesel engine which uses a catalyst to convert NO in the exhaust gas to NO<sub>2</sub>, thereafter trapping particulate in a filter and oxidising the particulate by reaction with the NO<sub>2</sub>. The exhaust gas is thereafter fed to a NO<sub>x</sub> absorbent or, in one embodiment, to a NO<sub>x</sub> conversion catalyst. Unburnt hydrocarbons and CO in the exhaust gas are trapped in a zeolite and released to react with NO<sub>x</sub> on the NO<sub>x</sub> conversion catalyst.

15 The technique named SCR (Selective Catalytic Reduction) is well established for industrial plant combustion gases, and may be broadly described as passing a hot exhaust gas over a catalyst in the presence of a nitrogenous reductant, especially ammonia or urea. This is effective to reduce the NO<sub>x</sub> content of the exhaust gases by about 20-25% at about 250°C, or possibly rather higher using a platinum catalyst, although platinum catalysts tend to oxidise NH<sub>3</sub> to NO<sub>x</sub> during higher temperature operation. We believe that SCR systems  
20 have been proposed for NO<sub>x</sub> reduction for vehicle engine exhausts, especially large or heavy duty diesel engines, but this does require on-board storage of such reductants, and is not believed to have met with commercial acceptability at this time.

25 We believe that if there could be a significant improvement in performance of SCR systems, they would find wider usage and may be introduced into vehicular applications. It is an aim of the present invention to improve significantly the conversion of NO<sub>x</sub> in a SCR system, and to improve the control of other pollutants using a SCR system.

30 Accordingly, the present invention provides an improved SCR catalyst system for treating combustion exhaust gas containing NO and particulates, comprising in combination and in order, an oxidation catalyst effective to convert NO to NO<sub>2</sub> and enhance the NO<sub>2</sub>

content of the exhaust gas, a particulate filter, a source of reductant fluid, injection means for said reductant fluid located downstream of said particulate trap and an SCR catalyst.

The invention further provides an improved method of reducing pollutants, including particulates and NO<sub>x</sub> in gas streams, comprising passing such gas stream over an oxidation catalyst under conditions effective to convert at least a portion of NO in the gas stream to NO<sub>2</sub>, and enhance the NO<sub>2</sub> content of the gas stream, removing at least a portion of said particulates in a particulate trap, reacting trapped particulate with NO<sub>2</sub>, adding reductant fluid to the gas stream to form a gas mixture downstream of said trap, and passing the gas mixture over an SCR catalyst.

Although the present invention provides, at least in its preferred embodiments, the opportunity to reduce very significantly the NO<sub>x</sub> emissions from the lean (high in oxygen) exhaust gases from diesel and similar engines, it is to be noted that the invention also permits very good reductions in the levels of other regulated pollutants, especially hydrocarbons and particulates.

The invention is believed to have particular application to the exhausts from heavy duty diesel engines, especially vehicle engines, eg truck or bus engines, but is not to be regarded as being limited thereto. Other applications might be LDD (light duty diesel), GDI, CNG (compressed natural gas) engines, ships or stationary sources. For simplicity, however, the majority of this description concerns such vehicle engines.

We have surprisingly found that a "pre-oxidising" step, which is not generally considered necessary because of the low content of CO and unburnt fuel in diesel exhausts, is particularly effective in increasing the conversion of NO<sub>x</sub> to N<sub>2</sub> by the SCR system. We also believe that minimising the levels of hydrocarbons in the gases may assist in the conversion of NO to NO<sub>2</sub>. This may be achieved catalytically and/or by engine design or management. Desirably, the NO<sub>2</sub>/NO ratio is adjusted according to the present invention to the most beneficial such ratio for the particular SCR catalyst and CO and hydrocarbons are oxidized prior to the SCR catalyst. Thus, our preliminary results indicate that for a transition



metal/zeolite SCR catalyst it is desirable to convert all NO to NO<sub>2</sub>, whereas for a rare earth-based SCR catalyst, a high ratio is desirable providing there is some NO, and for other transition metal-based catalysts gas mixtures are notably better than either substantially only NO or NO<sub>2</sub>. Even more surprisingly, the incorporation of a particulate filter permits still  
5 higher conversions of NOx.

The oxidation catalyst may be any suitable catalyst, and is generally available to those skilled in art. For example, a Pt catalyst deposited upon a ceramic or metal through-flow honeycomb support is particularly suitable. Suitable catalysts are e.g. Pt/Al<sub>2</sub>O<sub>3</sub>  
10 catalysts, containing 1-150g Pt/ft<sup>3</sup> (0.035-5.3g Pt/litre) catalyst volume depending on the NO<sub>2</sub>/NO ratio required. Such catalysts may contain other components providing there is a beneficial effect or at least no significant adverse effect.

The source of reductant fluid conveniently uses existing technology to inject fluid  
15 into the gas stream. For example, in the tests for the present invention, a mass controller was used to control supply of compressed NH<sub>3</sub>, which was injected through an annular injector ring mounted in the exhaust pipe. The injector ring had a plurality of injection ports arranged around its periphery. A conventional diesel fuel injection system including pump and injector nozzle has been used to inject urea by the present applicants. A stream of  
20 compressed air was also injected around the nozzle; this provided good mixing and cooling.

The reductant fluid is suitably NH<sub>3</sub>, but other reductant fluids including urea, ammonium carbamate and hydrocarbons including diesel fuel may also be considered. Diesel fuel is, of course, carried on board a diesel-powered vehicle, but diesel fuel itself is  
25 a less selective reductant than NH<sub>3</sub> and is presently not preferred.

Suitable SCR catalysts are available in the art and include Cu-based and vanadia-based catalysts. A preferred catalyst at present is a V<sub>2</sub>O<sub>5</sub>/WO<sub>3</sub>/TiO<sub>2</sub> catalyst, supported on a honeycomb through-flow support. Although such a catalyst has shown good performance  
30 in the tests described hereafter and is commercially available, we have found that sustained high temperature operation can cause catalyst deactivation. Heavy duty diesel engines,

which are almost exclusively turbocharged, can produce exhaust gases at greater than 500°C under conditions of high load and/or high speed, and such temperatures are sufficient to cause catalyst deactivation. In one embodiment of the invention, therefore, cooling means is provided upstream of the SCR catalyst. Cooling means may suitably be activated by  
5 sensing high catalyst temperatures or by other, less direct, means, such as determining conditions likely to lead to high catalyst temperatures. Suitable cooling means include water injection upstream of the SCR catalyst, or air injection, for example utilising the engine turbocharger to provide a stream of fresh intake air by-passing the engine. We have observed a loss of activity of the catalyst, however, using water injection, and air injection  
10 by modifying the turbocharger leads to higher space velocity over the catalyst which tends to reduce NOx conversion. Preferably, the preferred SCR catalyst is maintained at a temperature from 160°C to 450°C.

We believe that in its presently preferred embodiments, the present invention may  
15 depend upon an incomplete conversion of NO to NO<sub>2</sub>. Desirably, therefore, the oxidation catalyst, or the oxidation catalyst together with the particulate trap if used, yields a gas stream entering the SCR catalyst having a ratio of NO to NO<sub>2</sub> of from about 4:1 to about 1:3 by vol, for the commercial vanadia-type catalyst. As mentioned above, other SCR catalysts perform better with different NO/NO<sub>2</sub> ratios. We do not believe that it has previously been  
20 suggested to adjust the NO/NO<sub>2</sub> ratio in order to improve NOx reduction.

The present invention incorporates a particulate trap downstream of the oxidation catalyst. We discovered that soot-type particulates may be removed from a particulate trap by "combustion" at relatively low temperatures in the presence of NO<sub>2</sub>. In effect, the  
25 incorporation of such a particulate trap serves to clean the exhaust gas of particulates without causing accumulation, with resultant blockage or back-pressure problems, whilst simultaneously reducing a proportion of the NOx. Suitable particulate traps are generally available, and are desirably of the type known as wall-flow filters, generally manufactured from a ceramic, but other designs of particulate trap, including woven, knitted or non-woven  
30 heat-resistant fabrics, may be used.

It may be desirable to incorporate a clean-up catalyst downstream of the SCR catalyst, to remove any  $\text{NH}_3$  or derivatives thereof which could pass through unreacted or as by-products. Suitable clean-up catalysts are available to the skilled person.

5 A particularly interesting possibility arising from the present invention has especial application to light duty diesel engines (car and utility vehicles) and permits a significant reduction in volume and weight of the exhaust gas after-treatment system, in a suitable engineered system.

10 Several tests have been carried out in making the present invention. These are described below, and are supported by results shown in graphical form in the attached drawings.

15 A commercial 10 litre turbocharged heavy duty diesel engine on a test-bed was used for all the tests described herein.

### Test 1 - (Comparative)

20 A conventional SCR system using a commercial  $\text{V}_2\text{O}_5/\text{WO}_3/\text{TiO}_2$  catalyst, was adapted and fitted to the exhaust system of the engine.  $\text{NH}_3$  was injected upstream of the SCR catalyst at varying ratios. The  $\text{NH}_3$  was supplied from a cylinder of compressed gas and a conventional mass flow controller used to control the flow of  $\text{NH}_3$  gas to an experimental injection ring. The injection ring was a 10cm diameter annular ring provided with 20 small injection ports arranged to inject gas in the direction of the exhaust gas flow.

25  $\text{NO}_x$  conversions were determined by fitting a  $\text{NO}_x$  analyser before and after the SCR catalyst and are plotted against exhaust gas temperature in Figure 1. Temperatures were altered by maintaining the engine speed constant and altering the torque applied.

30 A number of tests were run at different quantities of  $\text{NH}_3$  injection, from 60% to 100% of theoretical, calculated at 1:1  $\text{NH}_3/\text{NO}$  and 4:3  $\text{NH}_3/\text{NO}_2$ . It can readily be seen that at low temperatures, corresponding to light load, conversions are about 25%, and the highest

conversions require stoichiometric (100%) addition of  $\text{NH}_3$  at catalyst temperatures of from 325 to 400°C, and reach about 90%. However, we have determined that at greater than about 70% of stoichiometric  $\text{NH}_3$  injection,  $\text{NH}_3$  slips through the SCR catalyst unreacted, and can cause further pollution problems.

5

## Test 2 (Comparative)

The test rig was modified by inserting into the exhaust pipe upstream of the  $\text{NH}_3$  injection, a commercial platinum oxidation catalyst of 10.5 inch diameter and 6 inch length (26.67cm diameter and 15.24cm length) containing 10g Pt/ft<sup>3</sup> (= 0.35g/litre) of catalyst volume. Identical tests were run, and it was observed from the results plotted in Figure 2, that even at 225°C, the conversion of NOx has increased from 25% to >60%. The greatest conversions were in excess of 95%. No slippage of  $\text{NH}_3$  was observed in this test nor in the following test.

15

## Test 3

The test rig was modified further, by inserting a particulate trap before the  $\text{NH}_3$  injection point, and the tests run again under the same conditions at 100%  $\text{NH}_3$  injection and a space velocity in the range 40,000 to 70,000 hr<sup>-1</sup> over the SCR catalyst. The results are plotted and shown in Figure 3. Surprisingly, there is a dramatic improvement in NOx conversion, to above 90% at 225°C, and reaching 100% at 350°C. Additionally, of course, the particulates which are the most visible pollutant from diesel engines, are also controlled.

## 25 Test 4

An R49 test with 80%  $\text{NH}_3$  injection was carried out over a V2O5/WO3/TiO2 SCR catalyst. This gave 67% particulate, 89% HC and 87% NOx conversion; the results are plotted in Figure 4.

30

7 10.01.00

Additionally tests have been carried out with a different diesel engine, and the excellent results illustrated in Test 3 and 4 above have been confirmed.

The results have been confirmed also for a non-vanadium SCR catalyst.

**CLAIMS**

1. An improved SCR system for treating combustion exhaust gas containing NO and particulates, comprising in combination and in order, an oxidation catalyst effective to  
5 convert and enhance the NO<sub>2</sub> content of the exhaust gas, a particulate trap, a source of reductant fluid, injections means for such reductant fluid located downstream of said particulate trap and an SCR catalyst.
2. An SCR system according to claim 1, wherein the reductant fluid is NH<sub>3</sub>.
- 10 3. An SCR system according to claim 1 or 2, wherein the oxidation catalyst is a platinum catalyst carried on a through-flow honeycomb support.
4. An SCR system according to claims 1,2 or 3 wherein the particulate filter is a wall-  
15 flow filter.
5. An SCR system according to any one of the preceding claims, comprising also means to cool gases upstream of the SCR catalyst.
- 20 6. An SCR system according to claim 5, comprising also control means such that said gas cooling means is activated only when a high SCR catalyst temperature is detected or conditions are determined that are expected to lead to high catalyst temperatures.
7. A diesel engine provided with an SCR system according to any one of claims 1 to 5.
- 25 8. A light duty diesel engine according to claim 6, wherein the volume of the exhaust gas after-treatment system is reduced.
9. A method of reducing pollutants, including particulates and NO<sub>x</sub>, in gas streams,  
30 comprising passing such gas stream over an oxidation catalyst under conditions effective to convert at least a portion of NO in the gas stream to NO<sub>2</sub> and enhance the NO<sub>2</sub> content of the

gas stream, removing at least a portion of said particulates in a particulate trap, reacting trapped particulate with  $\text{NO}_2$ , adding reductant fluid to the gas stream to form a gas mixture downstream of said trap, and passing the gas mixture over an SCR catalyst under  $\text{NO}_x$  reduction conditions.

5

10. A method according to claim 9, wherein said gas stream is the exhaust from a diesel, GDI or CNG engine.

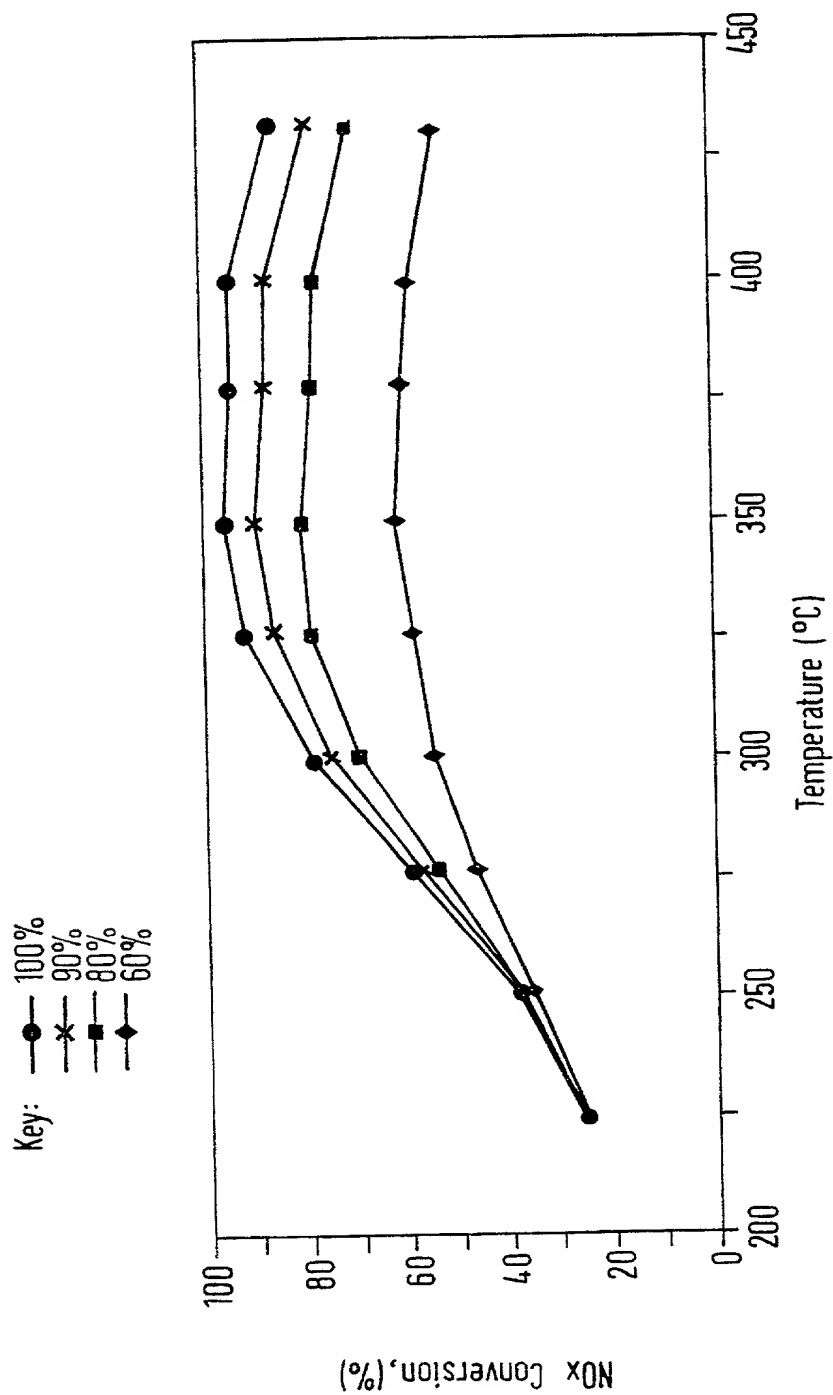
11. A method according to claim 9 or 10, wherein the gases are cooled, if necessary, before reaching the SCR catalyst.

10

12. A method according to claim 9, 10 or 11, wherein the  $\text{NO}$  to  $\text{NO}_2$  ratio in the gases is adjusted to a level pre-determined to be optimum for the SCR catalyst, by oxidation of  $\text{NO}$  over an oxidation catalyst.

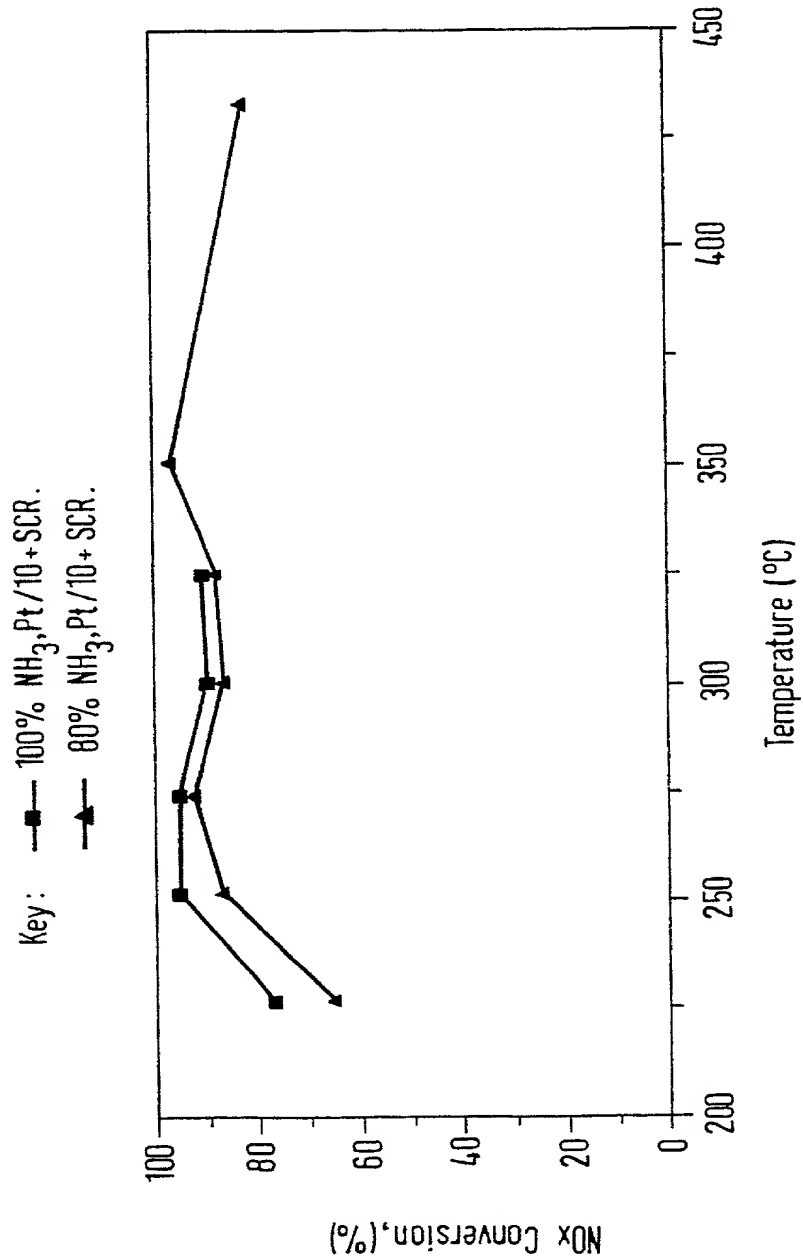
15

1/4

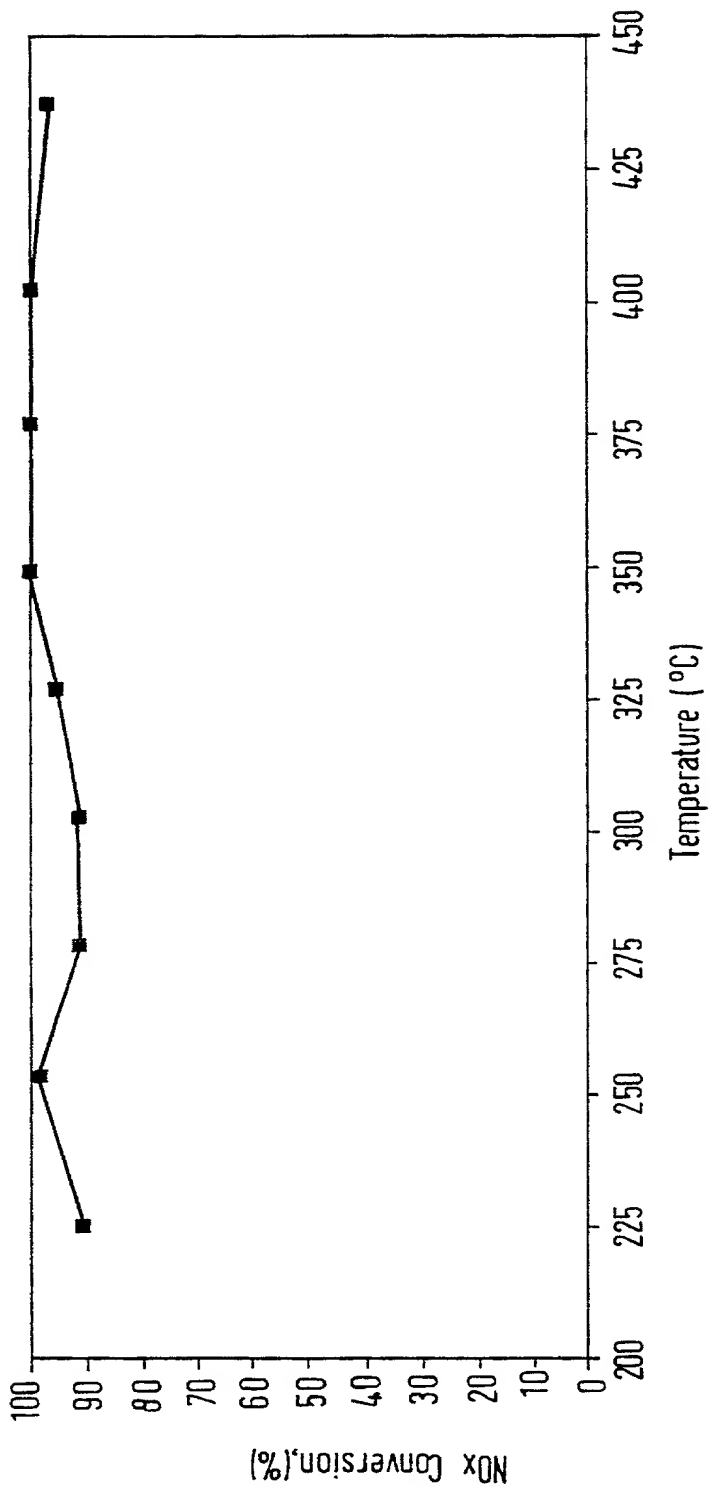




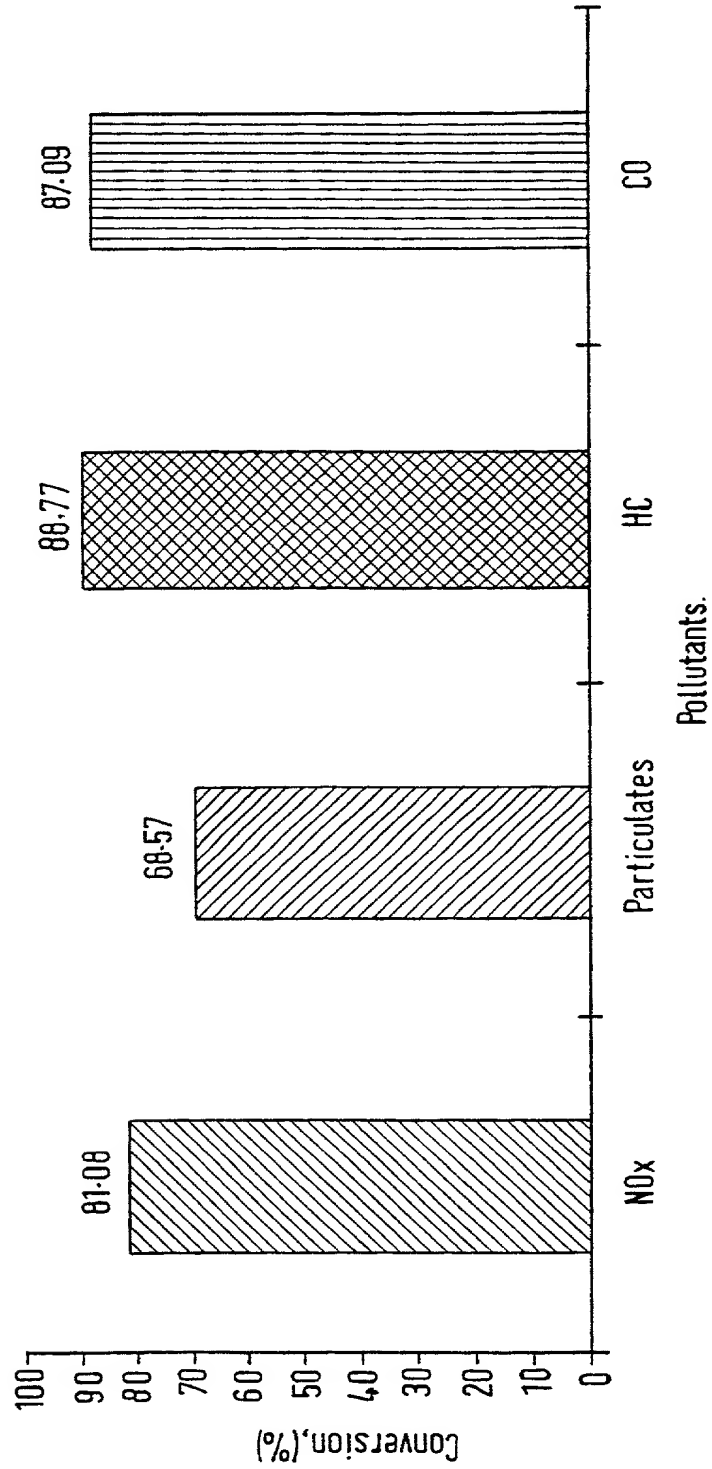
2/4



3/4



4/4



# Declaration and Power of Attorney For Patent Application

## English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

IMPROVEMENTS IN CATALYTIC REDUCTION OF NO<sub>x</sub>,

the specification of which is attached hereto unless the following box is checked:

☒ was filed on 28 January 1999 as  
United States Application Number or PCT International Application Number PCT/GB99/00292  
and was amended on 10 January 2000 and August 7, 2000 (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s) Priority Not Claimed

9802504.2 Great Britain 6 February 1998

(Number) (Country) (Day/Month/Year Filed) ☐

(Number) (Country) (Day/Month/Year Filed) ☐

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

(Application Number) (Filing Date)

(Application Number) (Filing Date)

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Number) (Filing Date) (Status - patented, pending, abandoned)

(Application Number) (Filing Date) (Status - patented, pending, abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Paul F. Prestia	Reg. No. <u>23,031</u>	Lawrence E. Ashery	Reg. No. <u>34,515</u>	Mark J. Marcelli	Reg. No. <u>36,593</u>
Allan Ratner	Reg. No. <u>19,717</u>	Christopher R. Lewis	Reg. No. <u>36,201</u>	Jack J. Jankovitz	Reg. No. <u>42,690</u>
Andrew L. Ney	Reg. No. <u>20,300</u>	Robert L. Andersen	Reg. No. <u>25,771</u>	Jonathan H. Spadt	Reg. No. <u>45,122</u>
Kenneth N. Nigon	Reg. No. <u>31,549</u>	Joshua L. Cohen	Reg. No. <u>38,040</u>	Christopher I. Halliday	Reg. No. <u>42,621</u>
Kevin R. Casey	Reg. No. <u>32,117</u>	Daniel N. Calder	Reg. No. <u>27,424</u>	Scott A. Mckeown	Reg. No. <u>42,866</u>
Benjamin E. Leace	Reg. No. <u>33,412</u>	Louis W. Beardell, Jr.	Reg. No. <u>40,506</u>		
James C. Simmons	Reg. No. <u>24,842</u>	Jacques L. Etkowicz	Reg. No. <u>41,738</u>		

Address all correspondence to: Paul F. Prestia

Ratner & Prestia, Suite 301, One Westlakes, Berwyn, P.O. Box 980, Valley Forge, PA 19482-0980

Address all telephone calls to: Paul F. Prestia at (610) 407-0700.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor (given name, family name) Anders ANDREASSON

Inventor's signature Anders Andreasson

Date 13/12-2000

Residence Traneredsvägen 60 A, S-426 77 V. Frolunda, Sweden

Citizenship Swedish

Post Office Address Traneredsvägen 60 A

S-426 77 V. Frolunda, SWEDEN

Full name of second joint inventor, if any (given name, family name) Guy Richard CHANDLER

Second inventor's signature G R Chandler

Date 14-11-00

Residence 48 Harlton Road, Little Eversden, Cambridge CB3 7HB, Great Britain

Citizenship British

Post Office Address 48 Harlton Road, Little Eversden

Cambridge CB3 7HB, GREAT BRITAIN



Additional inventors are being named on separately numbered sheets attached hereto.

3-0  
Full name of third joint inventor, if any (given name, family name) Claus Friedrich GOERSMANN

Third inventor's signature Claus Goermann Date 18-12-2000

Residence 39 Cromwell Road, Cambridge CB1 3EB, Great Britain GBN

Citizenship German

Post Office Address 39 Cromwell Road  
Cambridge CB1 3EB, GREAT BRITAIN

4-0  
Full name of fourth joint inventor, if any (given name, family name) James Patrick WARREN

Fourth inventor's signature James Patrick Warren Date 17-11-2000

Residence 67 Burnside, Cambridge CB1 3PA, Great Britain GBN

Citizenship British

Post Office Address 67 Burnside  
Cambridge CB1 3PA, GREAT BRITAIN

Full name of fifth joint inventor, if any (given name, family name) \_\_\_\_\_

Fifth inventor's signature \_\_\_\_\_ Date \_\_\_\_\_

Residence \_\_\_\_\_

Citizenship \_\_\_\_\_

Post Office Address \_\_\_\_\_

Full name of sixth joint inventor, if any (given name, family name) \_\_\_\_\_

Sixth inventor's signature \_\_\_\_\_ Date \_\_\_\_\_

Residence \_\_\_\_\_

Citizenship \_\_\_\_\_

Post Office Address \_\_\_\_\_

Full name of seventh joint inventor, if any (given name, family name) \_\_\_\_\_

Seventh inventor's signature \_\_\_\_\_ Date \_\_\_\_\_

Residence \_\_\_\_\_

Citizenship \_\_\_\_\_

Post Office Address \_\_\_\_\_